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THE EFFECT OF ENVIRONMENTAL FACTORS ON THE RESPIRATORY HEALTH OF SCHOOL CHILDREN IN HAMILTON

CURRENT REPORT #3 - APPENDICES

APRIL 1, 1980

URBAN AIR ENVIRONMENT GROUP

McMASTER UNIVERSITY

TO:

Health and Welfare Canada
Research Programs Directorate
Project 6606-1752-53

Ontario Ministry of the Environment
Air Resources Branch
Project 20

THE EFFECT OF ENVIRONMENTAL FACTORS ON THE RESPIRATORY HEALTH OF SCHOOL CHILDREN IN HAMILTON

CURRENT REPORT #3 - APPENDICES

APRIL 1, 1980

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APPENDIX 1-1

GLOSSARY OF TERMS

ADS - Anatomical Dead Space: The volume of the airways between the mouth and the alveoli, i.e., mouth, larynx, tracheal, and bronchial volumes.

CV - Closing capacity minus residual volume.

FEF_{25-75} - Forced Expiratory Flow: Mean forced expiratory flow between 25% and 75% points of FVC.

FEF_{75-85} - Forced Expiratory Flow: Mean forced expiratory flow between 75% and 85% of FVC.

FEV_t - Timed Forced Expiratory Volume: The volume of air expired in the time specified by t in seconds from the start of expiration, such as FEV_1 or FEV_3.

FRC - Functional Residual Capacity: The volume of air remaining in the lungs after a tidal expiration, or at the resting level (includes ADS).

FVC - Forced Vital Capacity: The maximum volume of air expired following a maximal inspiration, and using a maximal effort to expel the air.

IDI - Inspired Gas Distribution Index: An index of lung gas distribution found by dividing AAVV by 4.4 FRC (ideally, 4.38 FRC). The statistical mean value is 1.8 with a standard deviation of $\pm$0.2. An ideal one-compartment system would give an IDI of 1.00.

MEF_X – Maximal Expiratory Flow after X% of FVC has been exhaled.

MET - Time required to exhale from 25% to 75% of FVC.

APPENDIX 3-1

PROTOCOL AND CRITERIA TO BE USED BY THE PULMONARY FUNCTION TESTERS IN THE SCHOOLS

Year 2 - December 1979 (Unchanged from September 1979)

PROTOCOL

1. Each child is instructed before each test as to what is required.

2. The order of the tests is as follows. NWm x 1, FVC x 3, Spirometry x 1 and NWs x 2.

3. The tester ensures that the child is relaxed, that lips are sealed, and the noseclip is on properly.

4. NWm - This test is done with the child seated. Tidal breathing must be quiet, relaxed and regular when on the mouthpiece. When the child is ready the computer is switched in. The N2 must be falling; if not, the test will be invalid. The child continues to breathe quietly until less than 1% N2 is detected. The machine will stop automatically at about 0.8%. If the initial N2 readout on the computer display is less than 68% the test is invalid and must be repeated after a rest.

5. FVC - The child stands, non-supported (not leaning). The child breathes in room air to TLC, inserts FVC pneumotach, with a clean sterile mouthpiece into the mouth, and gives a forced expiration as rapidly

as possible to RV followed by a forced rapid inspiration back to TLC. The expiration must last for at least one second for the machine to record results.

6. Spirometry - This is done with the FVC pneumotach and the child seated. The child breathes quietly into the mouthpiece until the tester is satisfied with the relaxed VT, and switches in the computer. After four quiet VT breaths a maximum inspiration to TLC and expiration to RV is made. Quiet breathing is continued for three breaths to reestablish a stable FRC which should return to the pre VC baseline.

7. NWs - This is done seated with the N2 boom pneumotach. The child breathes normally (VT) until the machine signals ready then breathes out to RV. During expiration the O2 is switched on by the tester. At RV the child then inspires at a flow of 0.3 - 0.5 l/sec. to TLC and then expires at a similar flow rate to RV.

8. A short report is asked for all children. If parents have asked for results or if the child has low results (see criteria) a long report to the end of the FVC plot is asked for.

CRITERIA

GENERAL

Great care must be taken to ensure a good seal at the mouthpiece also that the child is relaxed and makes a maximum effort.

SPECIFICS

Nwm

Normal tidal breathing is essential. Test must be repeated if the initial N2 is less than 68% and if the final N2 is greater than 1%.

FVC

The two best VC's must be within 5% of each other. However, if the tester has done up to six attemps, with only one good VC resulting (the others are not within 5%), then the one good VC is still acceptable. If FVC/FEV1 ratio is less than 75%, then the three flow rates and also the MET sec are examined . Normal for the MET sec. is considered to be from .4 to .7. If the flow rates and the FVC/FEV1 are low, and the MET sec. is above .7 a long report is needed.

Spirometry

This VC is compared to the best FVC; if less than 10% the Spirometry is repeated, if more than 10%, the FVC is repeated. If ERV is less than 20% and more than 50% of VC, then the test must be repeated. FRC must be stable.

NWs

VC must be within 10% of largest previous VC. If VC greater than 10% of previous values repeat FVC and Spirometry. Two acceptable

manoeuvres should be obtained and up to five attempts made to obtain these. Nws Spirometry should liik okay; FRC must be stable and an obvious ERV and IC manoeuvre present (scale is too small to measure). Expired and inspired VC's (check NWs Spirometry) should be approximately the same.

COMMENTS

Children with colds or hayfever must have this data entered under "comments". Heavy bronchial lower chest colds are entered as "lower respiratory heavy", sneezing, dry coughing and upper chest colds are entered as "upper respiratory heavy", and sniffles as "slight cold".

Any medical information concerning the child and known to the tester e.g. asthma, should be entered also.

APPENDIX 3-2

YEAR 2 - December 1979 PROTOCOLS FOR THE DOCUMENTATION OF DATA FLOW

I MARKING I.D. LISTS

Mark on right hand side.
Mark all consents with tick.
If child moves after consent, put "m" after tick.
If child found to be ineligible after consent put "inel" after tick.
Mark all missing and unsigned consents with a "0".
If unsigned consent has moved or ineligible written on it, put "m" or "inel" after "0".
Mark refused consents with "Ø".
At end of list write in:- No. of consents with that I.D. No.
No. of consents from other schools.
Total consents.
Moved and ineligible children and subtract from total.
Final number to be tested.
Date by this list
When in school, subtract number of late movers and absentees.
This should then give final number tested.

I CONSENT BOOK

Mark the list at front of consent book the same way as the I.D. lists.
Enter totals, as above.
File I.D. numbers from other schools at the front.
File new kids, i.e. 300's, behind old kids.
File "Refused" consents at back of book.
Keep all moved, inel, etc. kids either with or without consent, loose at the back of the book and give them to Liz at the end of testing.

I AT END OF TESTING

Check coder's records with numbers in Liz's book.
Check these against totals on I.D. lists.
Write final numbers tested on school I.D. list, sign and date this.
Check consent book in similar manner.
Make sure all "movers" "inels" etc. are given to Liz.
Liz will enter numbers on flow sheets.

V AT END OF CODING

Check that numbers coded from coding record book, match numbers tested.
Write totals at end of coding I.D. list.
File both lists.
Check that diff I.D. numbers filed at beginning of book (after retests).
Check that all long reports for parents have been done.

APPENDIX 3-3

PROTOCOL TO BE USED IN OBTAINING INFORMATION OF HEALTH STATUS

1. On the day before testing, the assistant technician gives forms to the class teacher to give to the children. They are asked to take them home for their parents to fill in and to bring them back on the following day.

2. At the time of testing, information from the form is entered on a coding sheet. If the form is not returned, the code is 999.

3. Forms are marked and sorted into I.D. numbers and kept.

4. If a form comes in late and the coding sheets are still at the school, the assistant technician will update the information.

5. If the forms come in after the coding sheets have returned to McMaster, a list is made of the I.D. numbers. The information is subsequently updated on the computer by the data analyst.

6. At the end of testing within a school, all the forms are returned to McMaster and stored.

McMASTER UNIVERSITY
faculty of health sciences
1200 MAIN STREET WEST, HAMILTON, ONTARIO L8S 4J9

LETTER TAKEN HOME BY CHILD THE DAY BEFORE TESTING

Dear

We would be very grateful if you would fill in the following information and return this slip with your child tomorrow.

_____ has/had a cold (please check the appropriate box),

now . . . . . ☐
last week . . . . . ☐
2 weeks ago . . . . . ☐
3 weeks ago . . . . . ☐
4 weeks ago . . . . . ☐
5 to 6 weeks ago . . . . . ☐
more than 6 weeks ago . . . . . ☐

When he/she had this cold, did he/she also have a cough?

yes . . . . . ☐
no . . . . . ☐

If he/she has never had a cold, check this box ☐

Thank you for your help in giving us this information.

Yours sincerely

Anthony Kerigan

Dr. A. T. Kerigan
M.D., F.R.C.P.(C)
Scientific Coordinator

APPENDIX 3-4

PROTOCOL AND CRITERIA TO BE USED BY THE CODERS OF PULMONARY FUNCTION DATA

YEAR II - December 1979

PROTOCOL

1. The two assistant technicians code the Pulmonary Function Data. This is done in the schools immediately after each child is tested.

2. The Supervisor of Pulmonary Function Studies scrutinizes the coding of the NWs tests for both teams. This is done at McMaster in the week following testing.

3. When all coders have completed 20 tests, a randomized quality control is done.

CRITERIA

1. Patient Data - Information on the report must agree with the information slip for each child.

2. NWm - The initial N_2 concentration must not be less than 65% and must fall continuously to not more than 1%.

3. FVC - This must be within 10% of the biggest VC obtained on any test.

4. Spirometry - VC must be within 10% of biggest. FRC must be stable, i.e. the baseline must not deviate by more than 200 ml (by eye). FRC after VC maneuver should be approximately the same as before the maneuver. If the machine has chosen an incorrect FRC, the coders will correct this, recalculate the ERV and IC and adjust the RV and TLC values accordingly.

5. NWs - Procedure - The following variables will be recorded on Card 5: CV, N2DIFF, TLC, RV, FRC and Mean Flow. If there are two tests, both will be reported in full. The CV and N2DIFF values will be coded on Card 3.

If two tests meet the criteria, mean values are used.

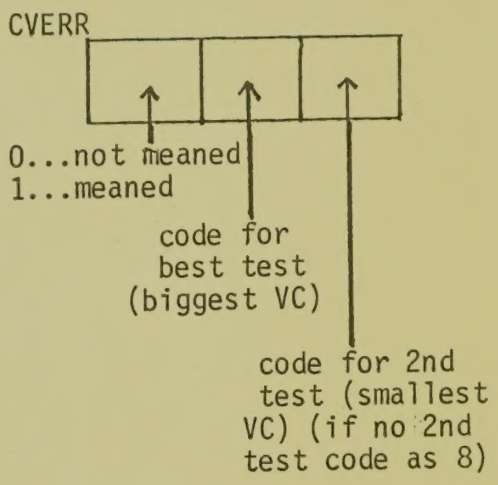
Criteria - The VC must be within 10% of the best VC. To mean two tests the VC's must be within 5% of each other. The Spirometry must look acceptable. (i.e. stable FRC, a visible ERV/IC maneuver). A correct IC/ERV ratio is vital to the calculation of FRC; therefore, if there is a falling/rising baseline and/or if there are no ERV/IC values printed and/or if the numbers for ERV and IC bear no relationship to the spirometry graph, the calculated FRV calue is invalidated. It is coded in these cases as 777.

APPENDIX 3-5

CODES NEW TO YEAR II - CARD 5

CODING OF ERRORS FOR NWs

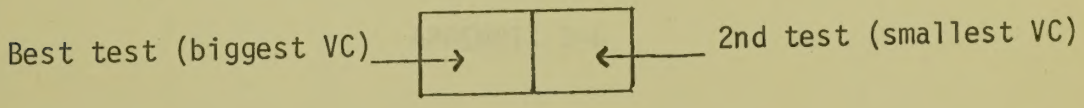
0 ... no error
1 ... wrong CV (start of slope of Phase IV)
2 ... wrong N2 DIFF (drawn)
3 ... wrong N2 DIFF (calculated)
4 ... combination 2/3
5 ... combination 1/2
6 ... combination 1/3
7 ... combination 1/2/3
8 ... not applicable
9 ... unknown



CODING OF CV & FRC AS BEING OF QUESTIONABLE VALIDITY

SUSPCV & SUSPFRC

(formula applies to both cases)

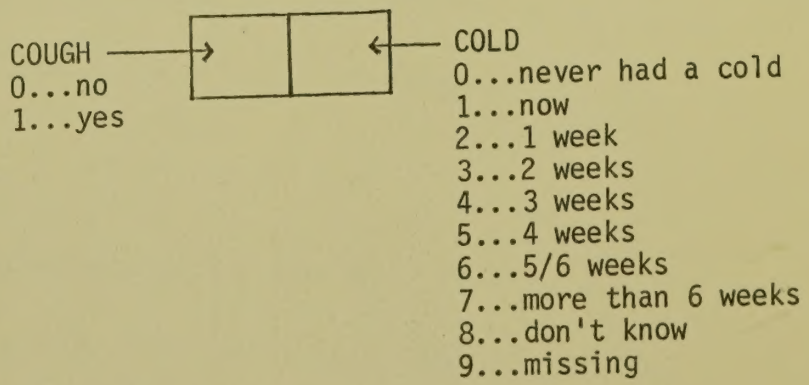


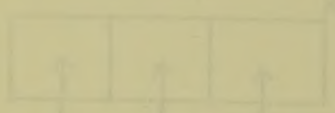
0 O.K. (no doubt as to acceptability)
1 Iffy (acceptable but required much subjective decision making)
7 Not acceptable

N.B. - Last year if results were unacceptable they were coded as 999.
This did not differentiate between missing or unobtainable data.

CODING OF INFORMATION RE CHILD'S HEALTH

"Cold When"





APPENDIX 3-6

SURVEY OF RESPIRATORY FUNCTION STATUS OF CHILDREN

I.D. [ ][ ][ ][ ][ ] (1–5)

CARD [3] (6)

ROOM [ ][ ][ ] (7–9)

AGE [ ][ ] (10–11)

WEIGHT [ ][ ][ ] (12–14)

SMOKER [ ][ ] (15–16)

TEMPERATURE [ ][ ] (17–18)

HUMIDITY % [ ][ ] (19–20)

SEX [ ] (21)

HEIGHT [ ][ ][ ] (22–24)

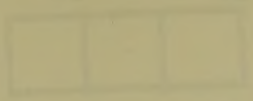
RACE/BIRTH [ ][ ][ ][ ][ ][ ][ ] (25–31)

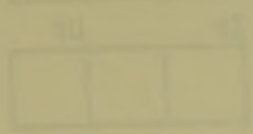
TESTER [ ] (32)

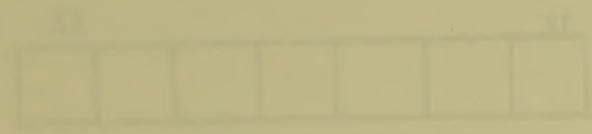
DATE/TIME [ ][ ][ ][ ][ ][ ][ ] (33–39)

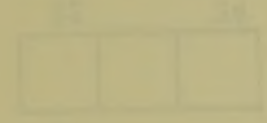
PRESSURE [ ][ ][ ] (40–42)

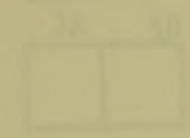
FVC [ ][ ][ ] (43–45)

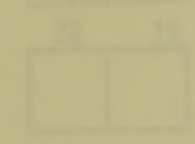


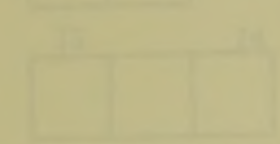


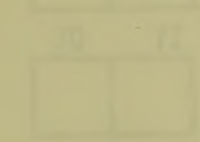


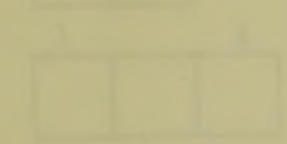


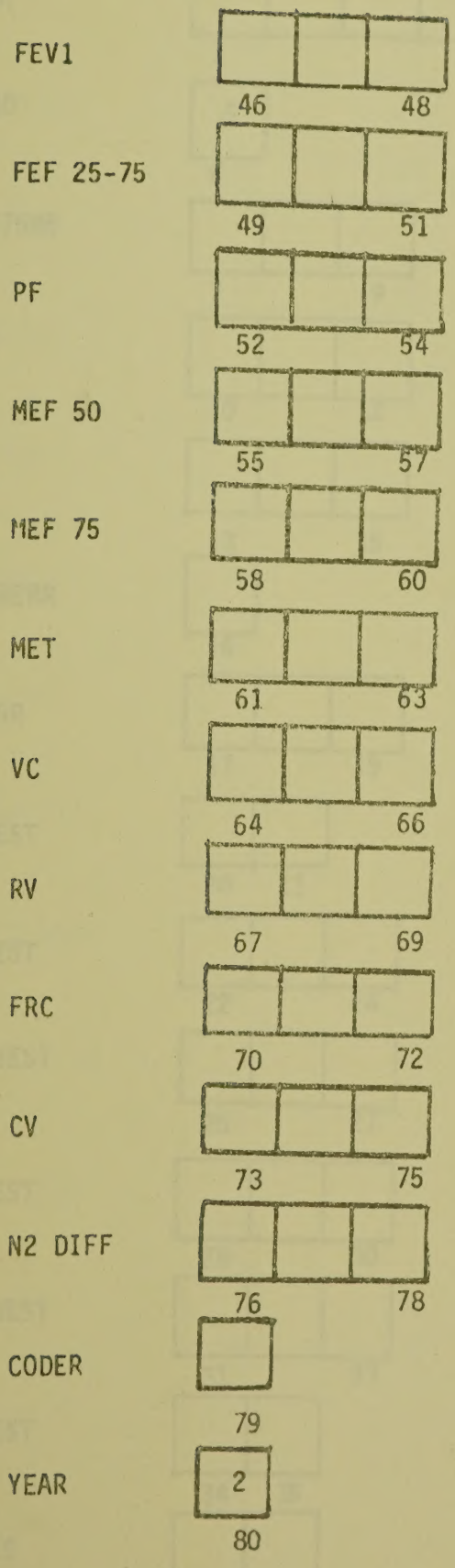











FEV1
46
48
FEF 25-75
49
51
PF
52
54
MEF 50
55
57
MEF 75
58
60
MET
61
63
VC
64
66
RV
67
69
FRC
70
72
CV
73
75
N2 DIFF
76
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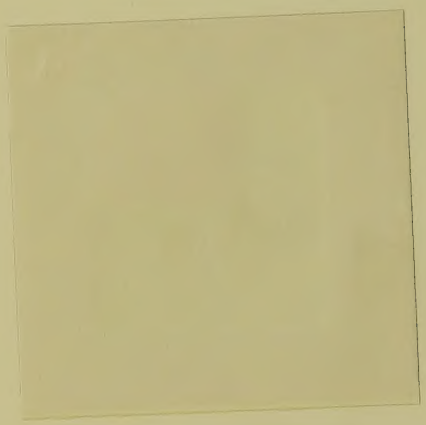
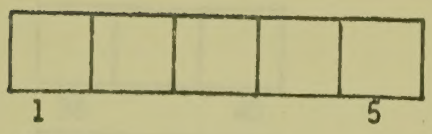
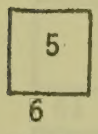
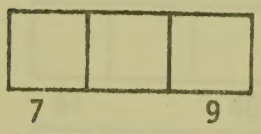
CODER
79
YEAR
2
80

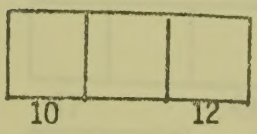


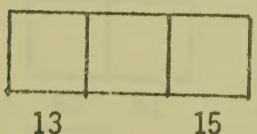
CARD 5 - Year 2

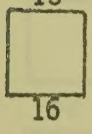
I.D. 

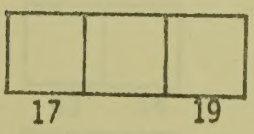
CARD 

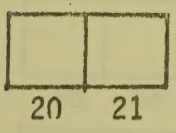
FEF7585 

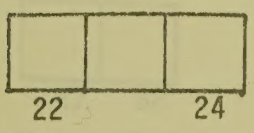
ADS 

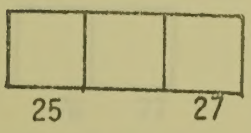
IDI 

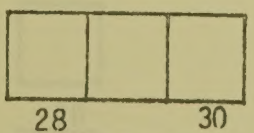
SPIRERR 

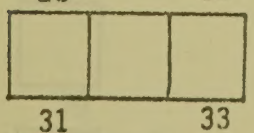
CVERR 

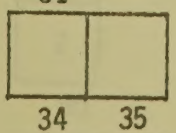
CVBEST 

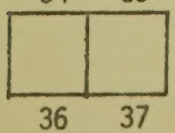
N2BEST 

TLCBEST 

RVBEST 

FRCBEST 

MFBEST 

CVSEC 

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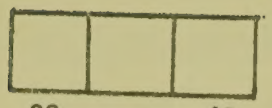

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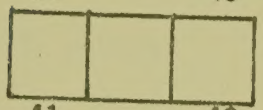

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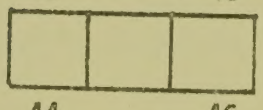

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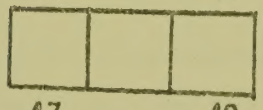

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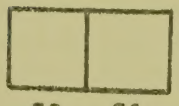

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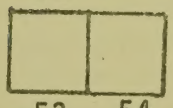

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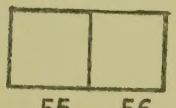

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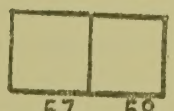

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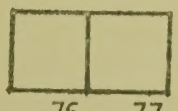

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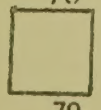

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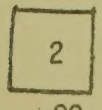

APPENDIX 6-1

Optical and Quantum Electronics 11 (*1979*) *385–391*

Detection of sulphur dioxide at, and below, the part per billion level using a tunable diode laser

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A laser absorption spectrometer is described which employs a wavelength tunable diode laser in conjunction with a multipass White cell. The instrument uses absorption lines in the ν_3 band of SO_2 to monitor atmospheric SO_2 concentrations with sensitivities as high as 0.2 parts per billion (ppb) in a laboratory system. A portable instrument with a fast response time and a sensitivity of 1 ppb is described, and the development of a highly practical field instrument, based on pyroelectric detectors, is discussed.

1. Introduction

In a series of previous papers, we describe the development of a laser absorption spectrometer (LAS) based on a tunable diode laser and a multipass White cell [1–3]. This instrument is sensitive to infra-red absorption coefficients as small as $3 \times 10^{-8}\,m^{-1}$ and has been exploited to develop a point monitoring system for numerous pollutants and trace gases of atmospheric significance. Much of our early work was aimed at the detection of SO_2 utilizing the ν_1 absorption band at $1150\,cm^{-1}$. Using a 5 m base path White cell and a 100 s time constant, we achieved a sensitivity of 1 part per billion (ppb) SO_2 in air [3]. We chose to operate initially in the $1150\,cm^{-1}$ spectral region despite the fact that the ν_1 absorption band is known to be much weaker than the ν_3 band in the region of $1350\,cm^{-1}$. However, the ν_3 band lies in a spectral region containing many strong water vapour absorption lines. As a consequence it seemed quite possible, at the time our work was initiated, that this multiplicity of strong absorption lines might interfere with the SO_2 absorption lines, and thereby negate the advantage of increased ν_3 absorption strength. Subsequent experience with the LAS system has shown that interferences from unwanted gaseous species in a gas sample are seldom a problem. Hence, we have switched to absorption lines in the ν_3 band for the detection of SO_2. As SO_2 is one of the most widespread gaseous pollutants in the atmosphere, we have carried out a series of experiments on the ν_3 band designed to improve the sensitivity and response time of the LAS for detection of this important gas.

The ν_3 band is by far the strongest infra-red band in SO_2. Lines in this band are approximately ten times as strong as those in the ν_1 band [4]. This increased line-strength has allowed us to replace the 5 m White cell with a short, commercial multipass cell, and to reduce the instrument time constant to 0.3 s, while retaining a sensitivity of 1 ppb SO_2 in air. No significant problems have been encountered with water vapour absorption, and we have achieved the order of magnitude improvement in detection sensitivity expected from the enhanced ν_3 band absorption. This increased detection capability can be used to reduce the instrument size and response time at a constant sensitivity, as indicated above. More importantly, it has allowed us to make another system trade-off of great practical significance. The LAS systems

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Detection of sulphur dioxide at, and below, the part per billion level using a tunable diode laser

used to date required highly sensitive HgCdTe detectors, which are both expensive and inconvenient from a practical system point of view, as their liquid nitrogen coolant requires regular replenishment. Our enhanced detection capability has permitted us to replace the HgCdTe detectors by inexpensive room-temperature, pyroelectric detectors, whilst maintaining adequate sensitivity. This development greatly improves the field deployability of the instrument, as it enables the entire apparatus to run unattended for days at a time. The present paper details the system improvements we have attained by switching to the ν_3 band of SO_2, and the trade-offs involved in changing from HgCdTe detectors to the more practical pyroelectric detectors.

2. Apparatus and results

2.1. Investigation of spectra of SO_2 and air

The apparatus employed is illustrated schematically in Fig. 1, and is described in detail in [1–3]. A beam splitter is used to split the output from the diode laser into two portions. The main beam is focused into the multipass White cell before falling on detector A. The gas to be monitored flows continuously at reduced pressure (~ 20 Torr) through this cell. The second laser beam is directed to detector B after passing through a short sample cell. This second beam can be used to lock the laser wavelength to the SO_2 absorption line under investigation. Further details are given later in this section. In our previous work on SO_2 monitoring we used a diode laser which tuned in the wavenumber region 1050–1180 cm^{-1}. For the present work this diode was replaced by one operating in the region 1200–1400 cm^{-1}. This diode was tuned into the ν_3 band of SO_2, which contains many strong lines between 1320 and 1390 cm^{-1}. The diode laser operated in a single mode over much of this band, with output powers of ~ 100 μW.

Fig. 2 shows some typical results obtained with this diode laser while examining the spectrum of pure SO_2, and SO_2 in air. For each trace, the laser is tuned in a single mode from 1349.7 cm^{-1} to 1350.3 cm^{-1} by ramping the diode laser current. In addition to this slow ramp, a small 1 kHz modulation is also superimposed on the diode laser current. The detector signal at 2 kHz is used as the Y input to the X–Y recorder, resulting in the second harmonic lineshapes illustrated in Fig. 2. (The advantages of the second harmonic technique for detecting very small absorptions are detailed in [1–3].) For the upper trace, a short cell containing pure SO_2 at ~ 5 Torr was placed in the laser beam. Several strong absorption lines were observed in this scan over 0.6 cm^{-1}. The experimental spectrum is compared in Fig. 2 with a theoretical spec-

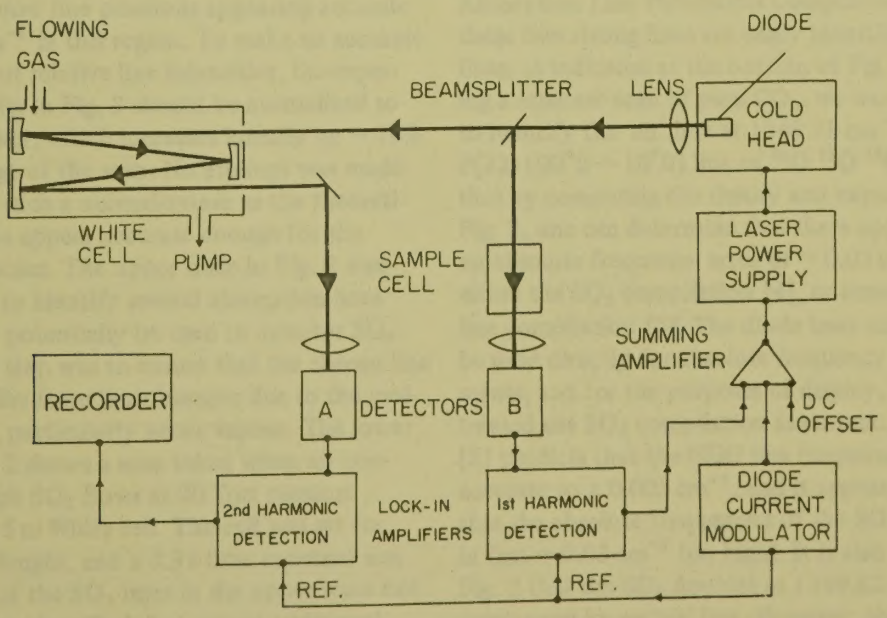


Figure 1 Schematic diagram of the laser absorption spectrometer (LAS).

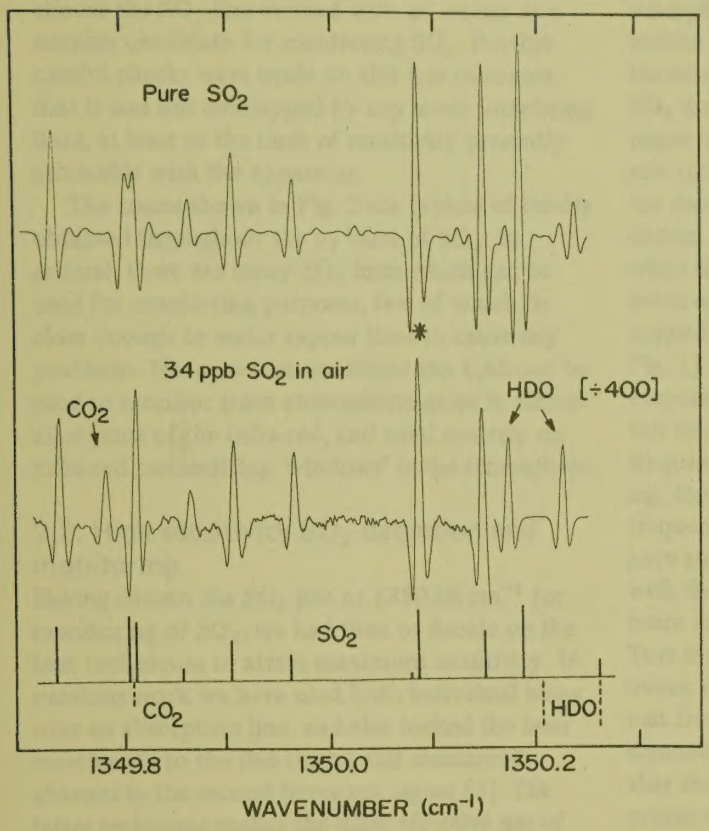


Figure 2 Spectra of pure SO_2, and SO_2 in air, taken near 1350 cm^{-1} using the tunable diode laser and second harmonic detection. For the upper trace a short cell containing pure SO_2 at 5 Torr was placed in the laser beam. The theoretical spectrum of SO_2 [4] in this region is indicated at the bottom of the figure. The lower trace shows a scan taken when air containing 34 ppb SO_2 flows at 20 Torr pressure through the 5 m White cell set for 160 m pathlength. Note the change in scale to display the strong HDO lines. The predicted positions of HDO and CO_2 lines [5] are also indicated at the bottom of the figure. The SO_2 line marked * is chosen to monitor SO_2. In this scan, it was recorded with a time constant of 0.3 s, and has an absorption coefficient of 10^{-5} m^{-1} (i.e. 3×10^{-7} m^{-1} ppb^{-1}).

trum taken from the AFGL trace gas compilation [4]. The overall agreement is reasonably good, with the *relative* line positions appearing accurate to ± 0.01 cm^{-1} in this region. To make an accurate comparison of relative line intensities, the experimental results in Fig. 2 should be normalized to the laser power, which increases linearly by ~ 70% over the range of the scan. No attempt was made to carry out such a normalization as the theoretical intensities appear accurate enough for the present purposes. The upper trace in Fig. 2 was simply used to identify several absorption lines which could potentially be used to monitor SO_2.

The next step was to ensure that the chosen line does not suffer from interferences due to the molecules in air, particularly water vapour. The lower trace in Fig. 2 shows a scan taken when air containing 34 ppb SO_2 flows at 20 Torr pressure through the 5 m White cell. The cell was set for 160 m path length, and a 0.3 s time constant was used. Many of the SO_2 lines in the upper trace can still clearly be identified, but several additional lines appear. In particular two very strong absorption lines appear near 1350.2 cm^{-1}. These two lines required a reduction in amplitude by a factor of 400 to display. Using the AFCRL Atmospheric Absorption Line Parameters Compilation [5], these two strong lines are easily identified as HDO lines, as indicated at the bottom of Fig. 2. By taking a separate scan of pure CO_2, we were also able to identify the 'air' line at 1349.78 cm^{-1} as the P(22) ($00^{\circ}0 \rightarrow 10^{\circ}0$) line of $^{16}O\ ^{12}O\ ^{18}O$. Note that by comparing the theory and experiment in Fig. 2, one can determine that there appears to be an absolute frequency error of ~ 0.03 cm^{-1} in either the SO_2 compilation [4], or atmospheric line compilation [5]. The diode laser scans cannot be used directly for absolute frequency measurements, and for the purposes of display, we have treated the SO_2 compilation as correct. (However, [5] predicts that the HDO line frequencies are accurate to ± 0.005 cm^{-1}, and it appears likely that the absolute frequencies of the SO_2 lines are in fact ~ 0.03 cm^{-1} too high). It is also clear from Fig. 2 that the SO_2 doublet at 1349.82 cm^{-1} is overlapped by an 'air' line. However, the molecule responsible for this line has not yet been identified. Nevertheless, based on Fig. 2 we were able to

choose the SO_2 line marked with an asterix as a suitable candidate for monitoring SO_2. Further careful checks were made on this line to ensure that it was not overlapped by any weak interfering lines, at least to the limit of sensitivity presently attainable with the apparatus.

The traces shown in Fig. 2 are typical of results obtained throughout the ν_3 band of SO_2. In general, there are many SO_2 lines which can be used for monitoring purposes, few of which lie close enough to water vapour lines to cause any problems. Hence we are confident the LAS can be used to monitor trace atmospheric gases in almost all regions of the infra-red, and need not rely on infra-red transmitting 'windows' in the atmosphere.

2.2. High sensitivity SO_2 detection and monitoring

Having chosen the SO_2 line at 1350.08 cm^{-1} for monitoring of SO_2, we had then to decide on the best techniques to attain maximum sensitivity. In previous work we have used both individual scans over an absorption line, and also locked the laser wavelength to the line centre and monitored changes in the second harmonic signal [3]. The latter technique makes the most effective use of the measurement time available, but is somewhat susceptible to baseline drifts, and d.c. drifts in the lock-in amplifier. For the present work, we have therefore chosen to use a repetitive scan over the SO_2 line for detection and monitoring. The technique is illustrated in Fig. 3. An additional small saw-tooth ramp (waveform 1) is superimposed on the diode current. The amplitude of this ramp is chosen so as to scan the laser over the SO_2 line, while the frequency of the ramp is low enough to avoid excessive distortion of the lineshape. The applied ramp and the output from detector A (see Fig. 1) are indicated in the left of Fig. 3. For most purposes, the diode laser is stable enough to run in this mode for a few hours without any appreciable frequency drift. However, for long term monitoring, the laser may drift away from the exact centre frequency of the SO_2 line. To prevent this, we have also operated the repetitive scan technique with the laser *locked* using the secondary laser beam and detector B. An SO_2/N_2 mixture at 50 Torr is placed in the sample cell, and first harmonic detection is used with detector B. The signals from detectors A and B as the diode current is scanned are compared on the right of Fig. 3. Note that the first derivative signal from the higher pressure cell is sufficiently broad that locking can be accomplished anywhere from frequency i to f

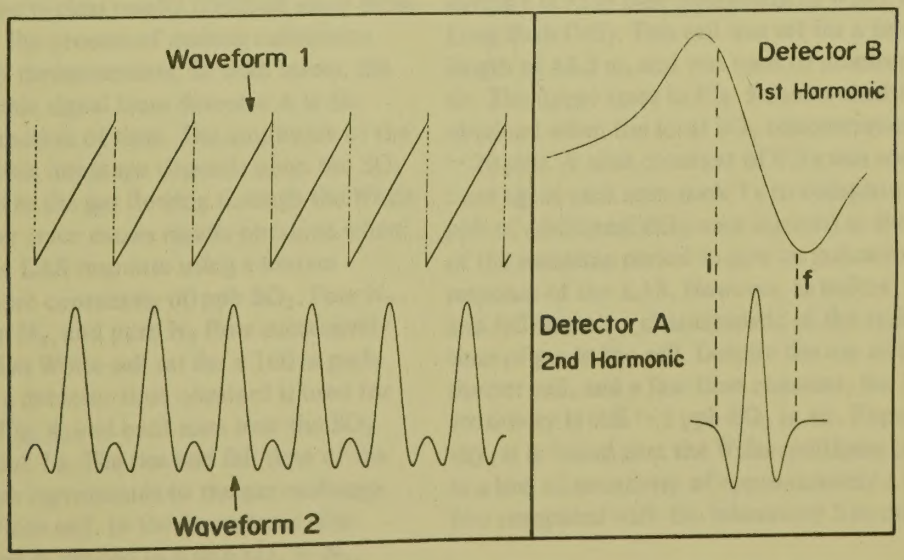
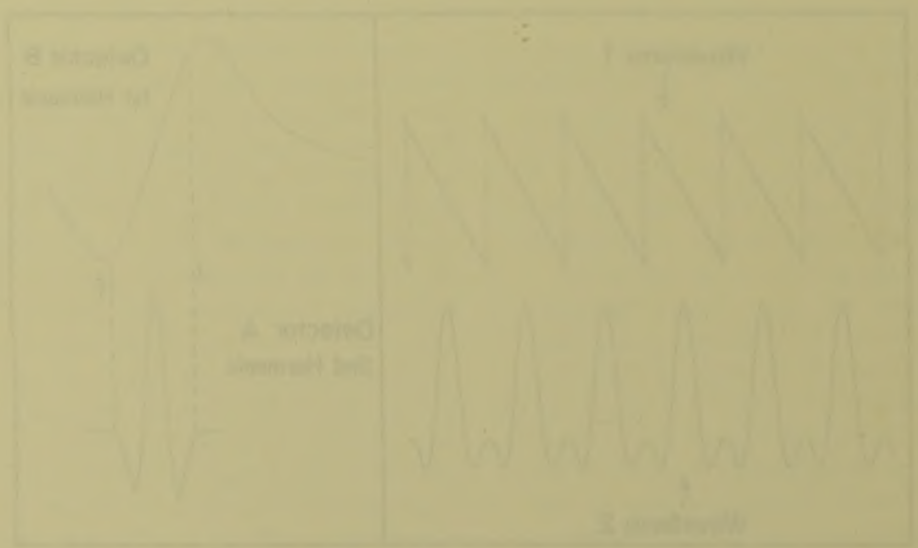


Figure 3 Illustration of the techniques used for continuous monitoring of SO_2. In addition to the 1 kHz modulation, a small saw-tooth ramp is superimposed on the diode current (waveform 1). This scans the laser repetitively over the SO_2 line, and results in the second harmonic waveform 2 at detector A. A similar repetitive scan can also be obtained with the laser under lock using detector B. The two traces on the right of Fig. 2 compare the second harmonic response from detector A with the first harmonic response from detector B. The laser can be locked and scanned over the wavelength range i to f. Details are given in the text.



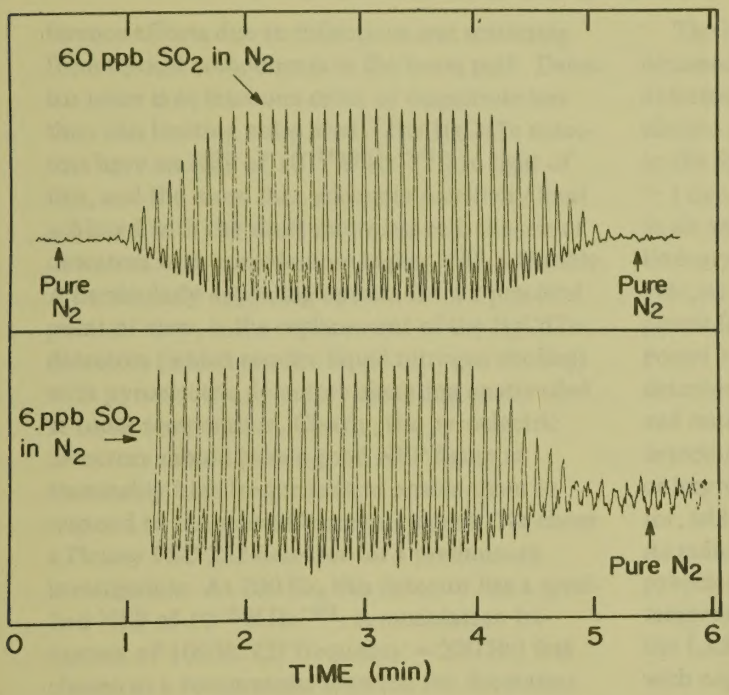


Figure 4 Calibration and sensitivity measurements on the LAS. The second harmonic signal from detector A is displayed as a function of time. The gas flowing through the White cell is also varied with time. In the upper trace, the instrument response was calibrated using a known 60 ppb SO_2 in N_2 mixture. For the lower trace, the lock-in amplifier gain was increased X 10, and the mixture diluted with N_2 to ~ 6 ppb. All results were obtained with 160 m pathlength in the 5 m White cell, and a time constant of 0.3 s.

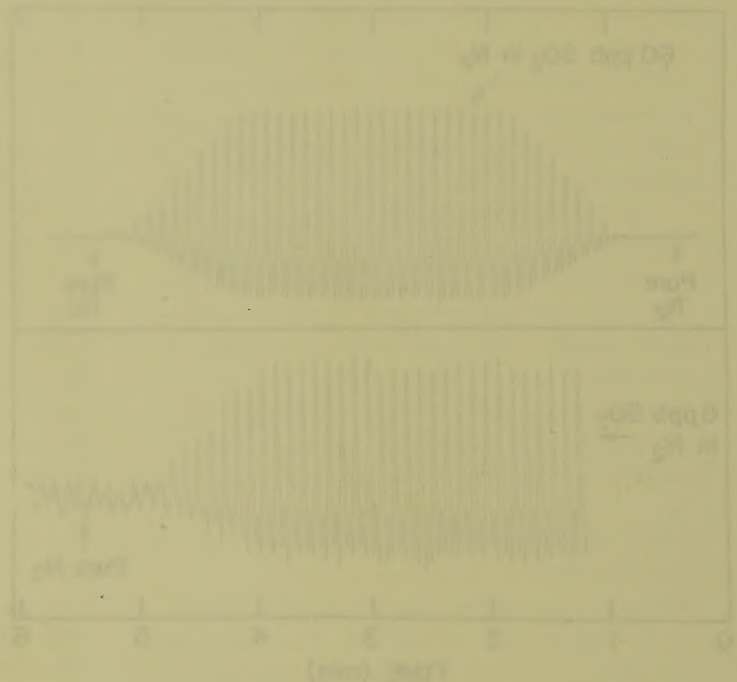
using a suitable reference voltage. A combination of conventional servo techniques and a saw-tooth offset voltage can thus be used to repetitively scan the SO_2 line under lock.

Fig. 4 shows typical results obtained using these techniques in the process of making calibration and sensitivity measurements. In both traces, the second harmonic signal from detector A is displayed as a function of time. The amplitude of the second harmonic lineshape depends upon the SO_2 concentration in the gas flowing through the White cell. The upper trace shows results obtained when calibrating the LAS response using a known SO_2/N_2 mixture containing 60 ppb SO_2. Pure N_2, 60 ppb SO_2 in N_2, and pure N_2 flow successively through the 5 m White cell set for a 160 m pathlength. A 0.3 s detector time constant is used for the traces in Fig. 4, and each scan over the SO_2 line takes about 7 s. The rise and fall time of the signal envelope corresponds to the gas exchange time in the White cell. In the lower trace, the known mixture is diluted to 6 ppb SO_2 in N_2, and from this trace we estimate the sensitivity as ~ 0.5 ppb. By increasing the time constant to 3 s, we have achieved detection limits for SO_2 of as little as 0.2 ppb [$(S/N) \sim 1$], using the long 5 m cell.

However, the main thrust of the present series of experiments was to develop a portable system with high sensitivity. We therefore replaced the 5 m White cell with a commercial multipass cell having a 0.75 m base pathlength (a Wilks Variable Long Path Cell). This cell was set for a total pathlength of 43.5 m, and was used to monitor SO_2 in air. The upper trace in Fig. 5 shows results obtained when the local SO_2 concentration was ~ 20 ppb. A time constant of 0.3 s was used, and once again each scan took 7 s to complete. A few ppb of additional SO_2 were injected in the middle of the sampling period to give an indication of the response of the LAS. However, as before, the rise and fall times are characteristic of the residence time of gas in the cell. Despite the use of the shorter cell, and a fast time constant, the LAS sensitivity is still ~ 1 ppb SO_2 in air. Experimentally, it is found that the Wilks multipass cell results in a loss of sensitivity of approximately a factor of two compared with the laboratory 5 m cell.

2.3. Use of pyroelectric detectors

Under all operating conditions of the LAS reported so far, the instrument sensitivity was limited by a combination of amplitude noise on the laser beam, and 'optical noise' caused by inter-



ference effects due to reflections and scattering from optical components in the beam path. Detector noise is at least one order of magnitude less than this limiting noise level. (The HgCdTe detectors have an NEP of 10^{-11} W $Hz^{-1/2}$.) In light of this, and the more than adequate sensitivity level achieved with the short multipass cell, the use of detectors with considerably higher NEP is feasible. A particularly appealing option, from a practical point of view, is the replacement of the HgCdTe detectors (which require liquid nitrogen cooling) with pyroelectric detectors operating unattended at room temperature. Clearly, the pyroelectric detectors should have a good NEP figure at reasonably high frequencies to enable them to respond to the second harmonic signals. We chose a Plessey PSC 222 detector for a preliminary investigation. At 200 Hz, this detector has a specified NEP of 10^{-9} W $Hz^{-1/2}$. A modulation frequency of 100 Hz (2f frequency = 200 Hz) was chosen as a compromise between the decreasing sensitivity of the pyroelectric at high frequencies, and the increasing amplitude noise on the laser beam at low frequencies. The pyroelectric detector was used with the lock-in amplifer time constant set at 3 s to limit the effect of detector noise.

The lower trace in Fig. 5 shows typical results obtained for monitoring SO_2 in air, when both detectors A and B have been replaced by pyroelectric detectors, and the LAS is operating locked to the SO_2 line. Each scan in the lower trace takes ~ 1 min to complete, and the SO_2 concentration in air was again 20 ppb. Sensitivity in these preliminary experiments was ~ 3 ppb SO_2 in air. However, no particular effort was made to optimize the power falling on the detectors. Of the 100 μW power emitted by the laser, only 20 μW falls on detector A after transmission through the lenses and multipass cell. As the sensitivity is limited by detector noise, any increase in transmitted power, or improvement in the NEP figure for the detector, will result directly in an increased sensitivity. As indicated earlier, a major advantage of using the pyroelectric detectors is that they operate at room temperature. We have therefore been able to run the LAS unattended for several days at a time, with negligible changes in alignment or sensitivity. Optimization of the laser power transmission and pyroelectric detector NEP should enable us to achieve long-term operation with sensitivities unchanged from those attained previously with the cooled HgCdTe detectors.

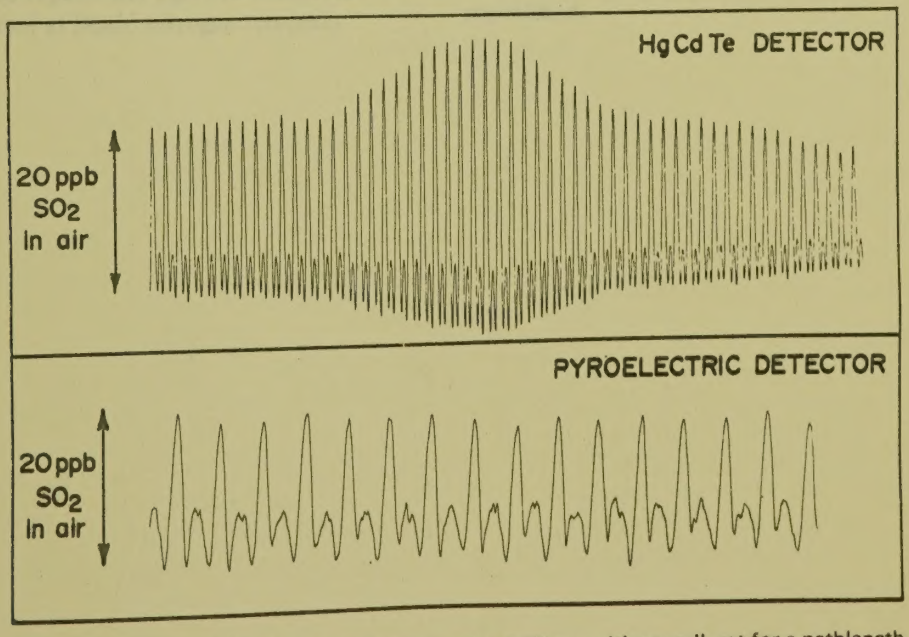


Figure 5 Continuous monitoring of SO_2 in air using the portable 0.75 m multipass cell, set for a pathlength of 43.5 m. The upper trace shows results obtained using HgCdTe detectors, a 0.3 s time constant, and took 7 min to complete. The lower trace was taken with the laser under lock, but used pyroelectric detectors in place of HgCdTe detectors A and B. Total scan time was 20 min, and a 3 s time constant was used to reduce the effect of detector noise. In each case, the local SO_2 concentration was ~ 20 ppb.

3. Discussion and conclusions

A point monitoring system has been described which employs a tunable diode laser and a multi-pass White cell to detect and monitor SO_2 in air. This system measures the absorption coefficients of SO_2 lines in the ν_3 band near 1350 cm^{-1}. The strength of these absorption lines is approximately an order of magnitude greater than those in the ν_1 band at 1150 cm^{-1}, which were used in our previous monitoring system. Despite the fact that the present system operates in a spectral region containing numerous strong water vapour lines, we show that it is immune from interferences from any of the molecules found in a typical urban atmosphere. Hence, the increased linestrength available in the ν_3 band can be translated directly into an order of magnitude improvement in system detection capability, over that previously achieved using the ν_1 band. This increased performance has been exploited in two ways. Firstly, the LAS has been operated with a small, commercially available White cell to produce a portable monitoring instrument with a sensitivity of 1 ppb and a fast response time. Secondly, and potentially even more important from a practical viewpoint, it has proved possible to replace the HgCdTe detectors (which are operated at liquid nitrogen temperatures) with lower sensitivity pyroelectric detectors, whilst maintaining adequate detection capability. The use of the room-temperature pyroelectric detectors enables the instrument to run unattended over long periods of time. We are in the process of developing a full portable instrument for evaluation under field conditions.

Acknowledgements

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APPENDIX 6-2

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gh sensitivity point monitoring of ozone, and high solution spectroscopy of the ν_3 band of ozone sing a tunable semiconductor diode laser

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Accurate measurements of the wavenumbers of more than 100 lines in the ν_3 band of ozone have been made using a tunable semiconductor diode laser and a temperature-stabilized germanium etalon. The accuracy for the line centers is estimated to be within 3–10 MHz and compares well with the accuracy attained using more complicated heterodyne techniques. The relevance of these results to ozone monitoring instruments based on CO_2 lasers is discussed. The tunability of the diode laser is also exploited to monitor ambient concentrations of ozone in the presence of interfering molecular species. A detection sensitivity of ~0.5 ppb of ozone is demonstrated.

Introduction

n recent years there has been a growing interest the measurement of ozone concentrations in the at-sphere. Stratospheric ozone plays an essential role he screening of harmful uv solar radiation from the th's surface. Consequently, the chemistry of ozone eing increasingly examined for possible ozone de-uction by man-made constituents such as Freon.[1] the other hand, ozone at the earth's surface is re-ded as a pollutant, because large concentrations of ne cause both irritation to humans and serious crop mage.[2] Reliable determination of ozone concentra-n requires the use of instrumentation having both h accuracy and sensitivity.

Ozone measurements employing lasers generally use sorption at the strong ν_3 ir band in the atmospheric ndow at 9 μm. The techniques include point and note sampling methods, using ground-based and borne instruments. Ground-based instruments ploying CO_2 lasers have been used with both passive d active targets to monitor ozone concentrations in e troposphere.[3,4] A similar airborne instrument has en used to monitor tropospheric ozone in the Los geles Basin.[4] In addition, ground-based solar het-odyne radiometers have been used to measure altitude ofiles of ozone in both the troposphere and strato-sphere.[5–7] All these instruments presently use CO_2 lasers, and all (especially the heterodyne radiometers) require accurate spectral data concerning the ozone line frequencies, linewidths, and line strengths. Ozone linewidths in the stratosphere have a half-width at half-maximum (HWHM) of approximately 30 MHz, and, therefore, their line-center frequencies must be known to better than 10 MHz if the heterodyne data are to be used properly. The entire ν_3 band of ozone has been analyzed by Barbe *et al.*[8] using measurements made on a SISAM interferometer. Their predicted accuracy ranges from ±30 MHz to ±100 MHz. A limited set of measurements has been made by Menzies[9] with an estimated accuracy of ±6 MHz. However, to date there has not been a comprehensive listing of the separation between O_3 lines and CO_2 laser lines, which has sufficiently high accuracy for design purposes in stratospheric ozone monitoring.

In the first part of this paper, we describe the accurate frequency measurements of over 100 lines in the ν_3 band of ozone. These measurements, taken with a laser absorption spectrometer (LAS) based on a tunable semiconductor diode laser,[10,11] are accurate within 3–10 MHz. Throughout this series of measurements, we concentrated on those ozone lines that are important for monitoring instruments based on CO_2 lasers. Hence, we have measured the frequencies of those reasonably strong ozone lines that lie within 6 GHz of the laser lines in the 9.4-μm region. These laser lines include most of the $^{12}C^{16}O_2$ lines and, in addition, selected isotopic and sequence lines,[12] particularly those in close coincidence (<300 MHz) with ozone lines. Diode lasers have been used previously to measure some individual absorption lines with accuracies of 15 MHz[13]

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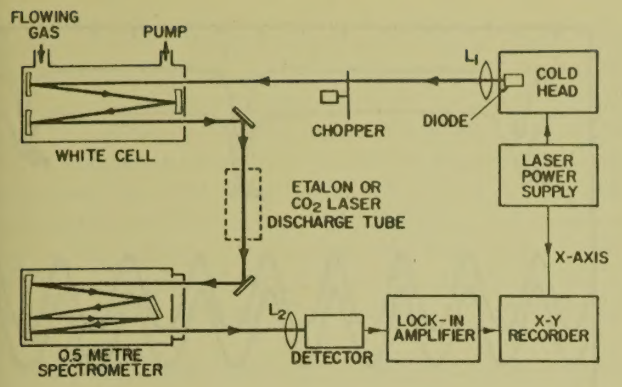


Fig. 1. Schematic diagram of the laser absorption spectrometer (LAS). L_1 and L_2 are the laser output and detector input lenses, respectively.

and to analyze absorption bands with much reduced accuracies.[14] However, this is one of the first reported measurements over an entire absorption band using the full resolution of the tunable diode laser.

Thus far we have discussed ozone detection based on CO_2 lasers. Such lasers are suitable if long pathlength propagation through the atmosphere is employed. For short pathlengths or for point monitoring of ozone, tunable diode lasers possess many advantages. For example, a tunable diode laser does not have to rely on chance coincidences between discrete laser lines and ozone absorption lines. An appropriate diode laser can be tuned to the line center of any strong ozone line and to a wavelength region that avoids interferences from other atmospheric species, particularly CO_2 and water vapor (instruments based on CO_2 lasers usually probe the atmosphere at several different frequencies in order to allow for interfering species).

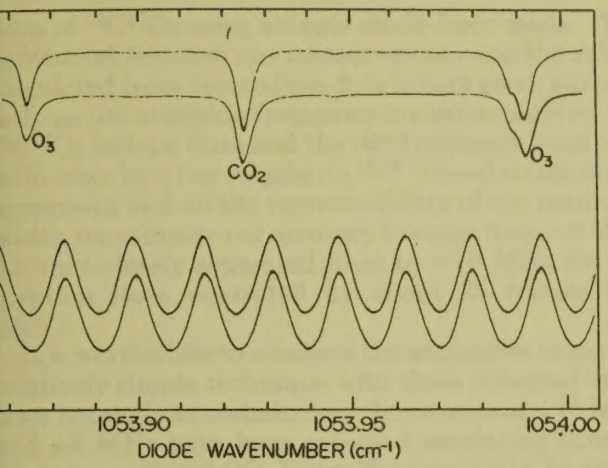
In the second part of this paper we report on the use of our LAS to measure ambient concentrations of ozone. We demonstrate a sensitivity of 0.5 ppb of ozone in the presence of all the interfering molecules normally found in an urban atmosphere. The LAS employed is sufficiently small that it can be built into a portable package suitable for either ground-based or airborne point monitoring of ozone.

II. Experimental Method

Figure 1 is a schematic diagram of the LAS. A PbSnSe diode (Laser Analytics, Incorporated) is mounted on the tip of a cold head, which is cooled by a closed-cycle He refrigerator. The diode temperature, which determines the approximate laser emission wavelength, can be set and controlled accurately in the 10–70-K range. Fine tuning of the wavelength is obtained by varying the diode current. A 1030–1070-cm^{-1} range was readily available from the diode laser used in our measurements. The laser output is collected by a 5-cm focal length lens, then chopped at 150 Hz, and finally focused into a 5-m base-pathlength multiple-traversal (White) cell. The output beam from the White cell passes through a 0.5-m spectrometer (to separate the longitudinal modes of the diode) and is then focused onto a HgCdTe detector. A phase-sensitive amplifier and an X–Y recorder are used to record the laser power as a function of diode current. As indicated in Fig. 1, a germanium etalon (described in Sec. III) can be inserted in the laser beam to provide a series of wavelength calibration fringes. There is also a provision for using a CO_2 laser discharge tube in the laser beam. This allows us to use gain measurements on selected CO_2 sequence lines, for example, as a method of obtaining additional reference lines. Further details of the apparatus are provided in Refs. 10 and 11.

We generally used an optical pathlength of 300 m in the White cell. Although not essential, such a long pathlength allows the use of low concentrations of ozone. Ozone, generated by flowing air through a simple uv ozone generator, was introduced into the White cell at a reduced pressure (~3 Torr). We estimate that the average ozone concentration in the cell was 10 ppm, based on the measured absorption of the strong ozone lines. The diode laser beam passes through this mixture of ozone and air in the White cell, through the 0.5-m scanning spectrometer, and finally onto the detector. The spectrometer serves two functions. Initially, it is used to select a single diode mode in the wavelength region of interest. For example, the spectrometer is set to pass ir radiation at 1053.9 cm^{-1} [i.e., the $P(12)$ CO_2 laser line]. The diode temperature and current are then varied until maximum detector output is achieved. Finally, a recording is made of absorption vs diode laser current in this wavelength region. The spectrometer now acts as a filter to reject any undesirable laser modes that may be present (these are generally separated by at least 2 cm^{-1} and, therefore, are easily rejected by a 0.5-m spectrometer).

It is easy to produce high-resolution recordings with a good diode laser. The main problem in high-resolution diode-laser spectroscopy is the accurate calibration of the wavelength scale. The usual procedure is to measure the separation between an unknown absorption line and a known reference line.[13,14] Fortunately, CO_2 provides numerous reference lines in the 9-μm wavelength region, and these have been measured with extremely high accuracy.[15] The upper traces in Fig. 2 show the $P(12)$ CO_2 reference line together with ozone lines obtained by adding a trace of CO_2 to the air–ozone mixture entering the White cell. The high sensitivity of our apparatus also enables us to use isotopic variations of CO_2 to provide additional reference lines. Both the $^{13}C^{16}O_2$ and $^{16}O^{12}C^{18}O$ forms of CO_2 can easily be detected in absorption using a 300-m pathlength in naturally occurring CO_2 (~5 Torr total pressure of CO_2). The frequencies of these isotopic laser lines have been measured to high accuracy by Freed *et al.*[16,17] In addition, we have employed a CO_2 discharge to generate the CO_2 sequence lines[18] as further references. As a result, we can generally find a CO_2 reference line within 0.4 cm^{-1} of any ozone line.



Typical example of data taken with the tunable diode laser. e upper scans, a mixture of 10-ppm ozone in air, together with e of CO_2, flows at ~3-Torr total pressure through the White cell n pathlength). The lower scans show reference fringes obtained with a germanium etalon.

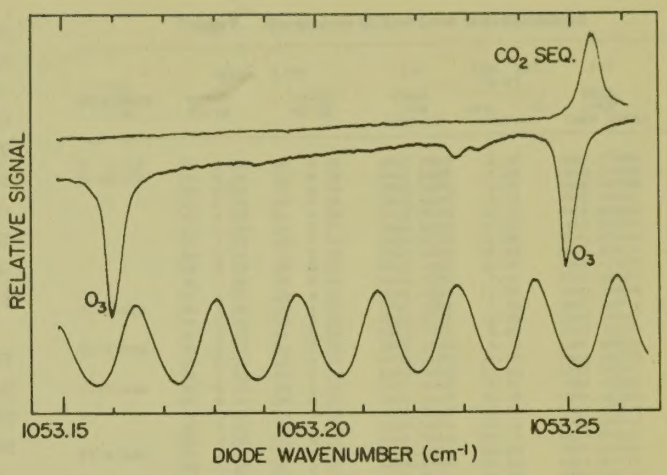


Fig. 3. Representative diode laser scans. For these, the diode laser beam is passed through a CO_2 discharge, through an ozone–air mixture in the White cell, and finally through the germanium etalon.

Table I. Measured Reference CO_2 Line Separations

CO2 LINES	(A) WAVENUMBER SEPARATION	NO. OF FRINGES (OUR WORK)	(B) WAVENUMBER SEPARATION (OUR WORK)	DIFF. (A)-(B) CONVERTED TO MHZ
P(28)636-P(30)SEQ	.00423 (REF.14,16)	.257	.00418	-2
P(20)626-P(31)628	.403112(REF.13,14)	24.888	.404960	-4
P(28)636-P(30)626	.267057(REF.13,14)	16.416	.267111	2
P(16)626-P(14)626	1.754263(REF.13)	107.813	1.754263	0

Ozone Wavenumbers

Accuracy of Measurements

ne final step in determining the accurate wave-ber of an ozone line, from data such as shown in Fig. to measure its separation from the reference CO_2

For this, we employed an uncoated solid germa-n etalon, nominally 7.650 cm long and with a finesse pproximately 3. The etalon was placed in the diode beam and provided calibration fringes as shown ne bottom of Fig. 2. The separation between a pair bsorption lines was measured by counting the ber of etalon fringes. Our aim was to measure the ne line frequencies to an accuracy of better than 10 z, which corresponds to less than two hundredths n etalon fringe. Care in etalon stabilization and ematic checking of the apparatus were, therefore, uired.

Ve first evaluated the stability of the laser, detector, X–Y recorder by comparing many successive traces ne type shown at the top of Fig. 2. Comparison of positions of the absorption lines on successive traces wed that the maximum shift of a line was less than Hz over a period of about an hour. Next, the etalon bility was checked. Initially, it was found that rmal changes cause the fringes to drift as much as a fringe (i.e., 240 MHz) from scan-to-scan. This esponds to a 0.08-K temperature change, based on culations using the data in Edwin *et al.*[19] Conse-ntly, the etalon was enclosed within a heavy copper nder whose temperature was stabilized by flowing er from a temperature controller through the cyl-er walls. This resulted in a random drift of ~0.05 fringes in periods of up to an hour, corresponding to the average etalon temperature remaining constant to better than 0.008 K. The very good reproducibility enabled us to measure absorption lines and then etalon fringes on successive scans with negligible loss of accuracy. In Fig. 3, we illustrate the scan of a reference CO_2 line in gain, an ozone absorption line, and the etalon fringes, using a single diode-laser beam. Note that the use of successive scans enables us to place the etalon in the diode-laser beam before it enters the spectrometer. Placing the etalon after the spectrometer in a two-beam system can lead to errors due to a changing angle of incidence at the etalon as the diode wavelength is changed.[13,20] A recent publication[21] describes a technique for the simultaneous recording of both the spectrum and the etalon fringes. This technique, which avoids errors due to changing angle of incidence, requires additional instrumentation but does not require such a high degree of stability from scan-to-scan.

Our usual measurement procedure was to take several recorder scans with ozone or CO_2 in the White cell, then several scans with the cell evacuated and the etalon in the beam. Finally, the initial scans were repeated as a check on stability. The scans were carefully measured by hand, with an estimated accuracy of 0.01–0.02 fringes (depending on the quality of the lines). This accuracy is based on the reproducibility of many successive measurements. To check for systematic errors, we made an initial series of measurements on CO_2 lines of known separation. The results of these measurements are given in Table I. The etalon fringe separation [0.016271(3) cm^{-1}] was calibrated by measuring the number of fringes between the $P(16)$ and $P(14)$ laser

ines of $^{12}C^{16}O_2$ using a single diode-laser mode. The greement between our measurements and the values alculated from heterodyne data is very good, particularly as the available frequency measurements on the $^{13}C^{16}O_2$ isotope lines and the 00°2 sequence lines may e in error by a few megahertz.[16,18] Based on the above greement and on the reproducibility of our measurements, we estimate our accuracy to range from ~3 MHz or very closely separated lines to ~10 MHz for absorption lines separated by about 25 fringes (0.4 cm^{-1}).

It is worthwhile to compare the accuracies using our relatively simple technique with those obtained using laser heterodyne techniques. Accuracies of ±15 MHz and ±3 MHz have been achieved employing tunable diode lasers[22] and CO_2 lasers,[23] respectively, to determine spectral line centers. The accuracy reported in this paper compares favorably with these values. Note that the heterodyne techniques are limited to measurements of absorption lines that lie within a detector bandwidth (~2 GHz) of the reference CO_2 lines.[22,23] No such limitation exists with our method.

At present, the accuracy of our measurements is still limited by residual drifts of the etalon fringes. Better temperature stabilization or the use of an air-spaced actively locked etalon will remove this limitation. The estimated accuracy of a tunable diode laser, in combination with a stabilized etalon, would then be limited simply by the accuracy with which one can judge the line center (1–3 MHz). Finally, it should be possible to automate much of the measurement procedure.

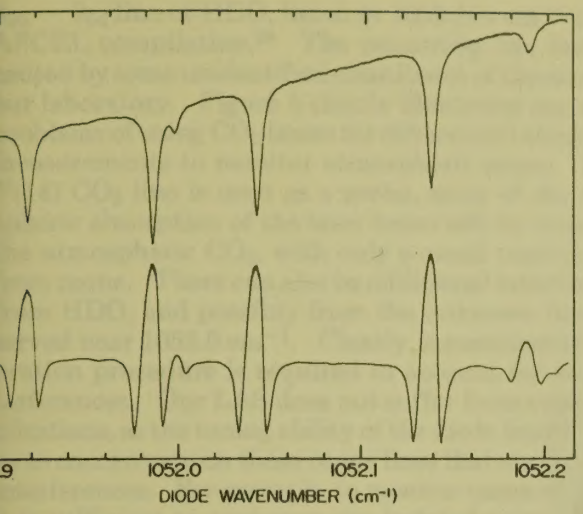
B. Results

The diode laser used in this work was operated in the 1030–1070-cm^{-1} range. By using regular and isotopic forms of CO_2 as references, we made frequency measurements of over 100 ozone lines in this wavenumber range, which is more than sufficient for a detailed band analysis of the ν_3 band of ozone; such an analysis is underway. Table II is a comprehensive list of relatively strong ozone lines that lie close to the important 9.4-μm laser lines of $^{12}C^{16}O_2$, to other CO_2 isotopes, and to the CO_2 sequence lines. These measured lines all lie within 0.4 cm^{-1} of the appropriate CO_2 lines and usually have intensities greater than 1×10^{-21} mole^{-1} cm^{-1}. We were able to measure approximately 55% of the lines satisfying these conditions at the listed CO_2 reference lines. The balance of the lines were either overlapped, or else the laser power was too low for carrying out accurate measurements. The listed line strengths are taken from Barbe *et al.*[8]; we did not undertake measurements of the line strengths. Our wavenumbers are in good agreement with Barbe *et al.* when their experimental uncertainties (up to 0.003 cm^{-1}) are taken into account.

In the regions covered by our diode laser, we found that all ozone lines with intensities greater than 1×10^{-21} mole^{-1} cm^{-1} (i.e., lines within 1/30th of the line strength of the strongest lines) could be identified with ν_3 lines listed by Barbe *et al.*[8] We found no evidence

Table II. Measured Ozone Line Wavenumbers

CO2 REFERENCE LINE	OZONE UPPER STATE	OZONE LOWER STATE	(A) CALC. 1/CM (REF.6)	ABS. INT. E-21 MOL. - CM (REF.6)	(B) MEAS. 1/CM (OUR WORK)	
R(4)626	39 3 36	38 3 35	1067.7369	8.18	1067.73480	
P(13)628	39 0 39	38 0 38	1062.6657	6.69	1062.66580	*
	38 8 31	37 8 30	1062.5998	2.49	1062.59998	
	36 7 30	35 7 29	1062.3995	4.85	1062.39836	
	32 4 29	31 4 28	1062.5818	10.53	1062.58266	
	32 3 30	31 3 29	1062.5778	11.82	1062.57773	
	33 1 32	32 1 31	1062.5313	11.86	1062.53142	
	34 2 33	33 2 32	1062.5252	10.63	1062.52614	
	29 2 27	28 2 26	1062.4932	16.39	1062.49392	
	40 9 32	39 9 31	1062.4224	1.44	1062.42053	
	31 4 27	30 4 26	1062.3895	11.56	1062.38968	
P(2)626	38 1 38	37 1 37	1062.3883	7.56	1062.38916	
	29 3 26	28 3 25	1062.2595	15.35	1062.25912	
	37 8 29	36 8 28	1062.1635	2.81	1062.16516	*
P(4)626	32 7 26	31 7 25	1060.6948	6.16	1060.69676	
	28 3 26	27 3 25	1060.6678	16.70	1060.66742	
	29 5 24	28 5 23	1060.5430	11.68	1060.54217	
P(17)628	27 5 22	26 5 21	1059.4495	13.79	1059.45203	
	29 0 29	28 0 28	1059.4497	18.49	1059.45283	
	25 1 24	24 1 23	1059.4327	22.84	1059.43514	
	26 4 23	25 4 22	1059.3880	17.12	1059.39043	*
P(6)626	28 6 23	27 6 22	1059.3682	10.79	1059.37069	
	23 2 21	22 2 20	1059.0351	24.54	1059.03409	
	28 1 28	27 1 27	1059.0109	19.92	1059.01115	
	25 4 21	24 4 20	1058.8898	18.19	1058.88916	
	26 5 22	25 5 21	1058.8828	14.73	1058.88153	
	30 8 23	29 8 22	1058.8669	5.72	1058.86700	
P(8)626	27 6 21	26 6 20	1058.8293	11.48	1058.82874	
	22 3 20	21 3 19	1057.3682	23.78	1057.36759	
	27 8 19	26 8 18	1057.3186	7.14	1057.31812	
	24 1 24	23 1 23	1057.2946	25.53	1057.29463	*
	31 10 21	30 10 20	1057.2293	2.27	1057.22781	
	22 2 21	21 2 20	1057.1907	25.99	1057.18928	
	23 5 18	22 5 17	1057.1631	17.32	1057.16175	
	24 6 19	23 6 18	1057.1537	13.69	1057.15467	
	34 11 24	33 11 23	1057.1308	1.75	1057.12953	
	22 4 19	21 4 18	1057.0529	21.09	1057.05130	
	21 3 18	20 3 17	1057.0421	24.66	1057.04049	
	25 7 18	24 7 17	1057.0050	10.41	1057.03416	
	23 0 23	22 0 22	1056.9442	26.85	1056.94258	
P(10)626	19 3 16	18 3 15	1055.7063	26.07	1055.70533	
	24 8 17	23 8 16	1055.6043	8.43	1055.60300	
	21 6 15	20 6 14	1055.3959	15.39	1055.39341	
	20 1 20	19 1 19	1055.3309	29.09	1055.33027	
	20 5 16	19 5 15	1055.3315	19.16	1055.33027	
	22 7 16	21 7 15	1055.3224	11.87	1055.32162	
	25 9 16	24 9 15	1055.3107	5.94	1055.30959	
P(12)626	25 10 15	24 10 14	1054.1945	4.15	1054.19462	
	19 6 13	18 6 12	1054.1729	16.92	1054.17213	
	20 7 14	19 7 13	1054.1303	12.48	1054.12951	
	18 5 14	17 5 13	1054.1103	16.99	1054.11298	
	17 0 17	16 0 16	1053.9664	31.66	1053.96577	
	21 8 13	20 8 12	1053.9606	9.33	1053.96025	
	17 4 13	16 4 12	1053.9203	23.30	1053.91944	*
P(24)628	17 5 12	16 5 11	1033.4586	19.84	1033.45478	*
P(9)SEQ	16 4 13	15 4 12	1033.2593	23.44	1033.25863	*
	22 12 11	21 12 10	1033.2498	1.33	1033.23969	
	26 11 16	25 11 15	1033.2418	2.45	1033.23570	
	16 1 16	15 1 15	1033.1683	31.55	1033.16756	
P(25)628	16 3 12	15 5 11	1032.8062	19.69	1032.80593	
	19 8 11	18 8 10	1032.7606	9.55	1032.75973	
	25 11 14	24 11 13	1032.7236	2.57	1032.71861	
	15 4 11	14 4 10	1032.5947	23.15	1032.59424	*
	22 10 13	21 10 12	1032.3618	4.62	1032.35940	
P(14)626	14 2 13	13 2 12	1032.4926	28.92	1032.39215	
	13 1 12	12 1 11	1032.3093	29.85	1032.30882	
	16 7 10	16 7 9	1032.2641	26.11	1032.26306	
	16 6 11	15 6 10	1032.2642	12.33	1032.26306	
	14 3 12	13 3 11	1032.2631	13.89	1032.26306	
	24 11 14	23 11 13	1032.1985	2.67	1032.19472	*
	18 8 11	17 8 10	1032.1439	9.52	1032.14361	
	15 5 10	14 5 9	1032.1438	19.83	1032.14361	
	13 2 11	12 2 10	1032.0436	20.17	1032.04254	
	21 10 11	20 10 10	1031.9984	4.70	1031.99600	
	14 1 14	13 1 13	1031.9836	30.96	1031.98448	
P(27)628	15 7 8	14 7 7	1030.9664	11.86	1030.96581	
	14 6 9	13 6 8	1030.9337	14.88	1030.93297	
	12 3 10	11 3 9	1030.8780	24.37	1030.87740	
	11 1 10	10 1 9	1030.8642	27.73	1030.86388	
	13 5 8	12 5 7	1030.7880	17.96	1030.78727	
P(16)626	12 1 12	11 1 11	1030.7412	29.27	1030.74042	
	17 9 8	16 9 7	1030.6560	6.63	1030.65565	
	11 2 9	10 2 8	1030.5436	25.93	1030.54453	
	12 4 9	11 4 8	1030.5329	20.00	1030.53194	
P(10)626	11 0 11	10 0 10	1030.3859	28.48	1030.38945	
	12 7 6	11 7 5	1040.9372	9.40	1040.93045	
	18 11 8	17 11 7	1040.9053	2.79	1040.90306	
	13 8 5	12 8 4	1040.8999	7.28	1040.89893	
	11 6 5	10 6 4	1040.8609	11.68	1040.86093	
	14 9 6	13 9 5	1040.7350	5.41	1040.73481	
	9 3 6	8 3 5	1040.7212	19.60	1040.72083	
P(28)626	10 5 6	9 5 5	1040.6742	13.93	1040.67385	*
	7 4 3	6 4 2	1046.8099	11.06	1046.80950	
	10 8 3	9 8 2	1046.8156	3.73	1046.81540	
	9 7 2	8 7 1	1046.8062	4.78	1046.80609	
	11 9 2	10 9 1	1046.7103	2.8	1046.71030	
	6 2 5	5 2 4	1046.6928	15.35	1046.69215	
	8 6 3	7 6 2	1046.6876	3.91	1046.68765	
	6 1 6	5 1 5	1046.5457	17.30	1046.64504	
R(44)636	5 4 1	4 4 0	1045.3567	4.42	1045.35660	*
P(22)626	4 1 4	3 1 3	1045.1643	11.72	1045.16408	
	4 2 3	3 2 2	1045.1408	8.96	1045.14060	
R(36)636	5 3 2	5 3 3	1041.4876	8.81	1041.48675	*
P(26)626	7 3 4	7 3 5	1041.3418	6.10	1041.34126	
	8 3 6	8 3 5	1041.2484	5.20	1041.24775	
P(30)626	16 8 9	16 8 8	1037.4342	5.50	1037.43452	*
R(28)636	4 3 2	5 3 3	1037.3419	8.55	1037.34157	
	20 7 14	20 7 13	1037.3217	3.13	1037.32131	
	12 9 4	12 9 3	1037.3132	9.07	1037.31297	
	23 6 17	23 6 16	1037.2395	1.06	1037.23780	
	17 8 9	17 8 10	1037.2318	4.83	1037.23133	
	13 9 4	13 9 5	1037.1478	7.94	1037.14746	
P(29)SEQ	7 1 6	8 1 7	1034.8563	21.69	1034.85533	
	7 2 5	8 2 6	1034.8250	19.82	1034.82405	
	14 11 4	14 11 3	1034.8173	3.16	1034.81621	
	29 7 22	29 7 23	1034.8603	.06	1034.80385	
	19 12 7	19 12 8	1034.7898	1.79	1034.78876	
	20 12 9	20 12 8	1034.7883	1.43	1034.78876	
	18 12 7	18 12 6	1034.7857	2.01	1034.78597	
	21 12 9	21 12 10	1034.7811	1.21	1034.78143	
	6 7 2	7 5 3	1034.7774	6.83	1034.77711	
	16 12 5	16 12 4	1034.7596	2.84	1034.76220	
	15 12 3	15 12 4	1034.7371	3.38	1034.73941	
	14 12 3	14 12 2	1034.7077	4.05	1034.71181	
	13 12 1	13 12 2	1034.6711	4.88	1034.67318	



High-resolution ozone spectrum near 1052 cm^{-1}. The upper as taken with conventional amplitude detection; second har- detection was employed for the lower scan. A mixture of n ozone in air was flowed through the White cell (240-m path- length) at a total pressure of 20 Torr.

ot band or isotope lines with significant line gth. This observation agrees with the results of e *et al.*[8]

e data in Table II are essential for comparing ex- nental and theoretical estimates of ozone absorp- as a function of pressure.[10,24] The wavenumbers s table are also sufficiently accurate to be used with heterodyne radiometers in the measurement of tropospheric and stratospheric ozone. Airborne uments employing differential absorption based O_2 lasers require a close coincidence between the e line and the CO_2 laser line. Such CO_2 lines, n are indicated by an asterisk in Table II, include us isotope and sequence lines. Despite the addi- l complexity in obtaining lasing on the rare isotope quence lines of CO_2, these may prove more practical ratospheric monitoring of ozone, as they suffer gible absorption by atmospheric CO_2.

Point Monitoring of Ozone

mentioned in Sec. I, the use of a tunable diode has several advantages compared with a CO_2 laser e point monitoring of ozone. To demonstrate this, ave used our diode laser and White cell to monitor ent levels of ozone in an urban atmosphere. The step in attaining high sensitivity detection of ozone determine an operating region in which the laser ates in a single mode and tunes through several g ozone lines. Single-mode operation enables the val of the spectrometer from the beam path, which ts in an improved SNR. We chose to operate the e laser in the 1052-cm^{-1} region near the $P(14)$ line O_2. The upper trace in Fig. 4 is a scan of a relatively concentration of ozone (~10 ppm), taken with the chopper in the laser beam, and employing conventional amplitude detection. The chopper was then removed from the system, and a small frequency modulation was superimposed on the diode current ramp. Next the detector signal at the second harmonic of this modulation frequency was recorded, as shown in the lower scan of Fig. 4. This second harmonic technique results in a substantial reduction in noise and enables us to detect absorption coefficients as low as 3×10^{-8} m^{-1} (see Refs. 10 and 11).

Ambient ozone levels in air are detected by scanning at high sensitivity. For our measurements, air from above the roof of the laboratory building was flowed through the White cell at a reduced pressure (20 Torr), and the diode laser was scanned over the same wavenumber region as in Fig. 4. The data are shown in Fig. 5, where four strong ozone lines can be clearly seen as well as several additional lines. The very strong line at 1052.196 cm^{-1} is easily identified as the $P(14)$ line of CO_2. One of the other additional lines is probably the

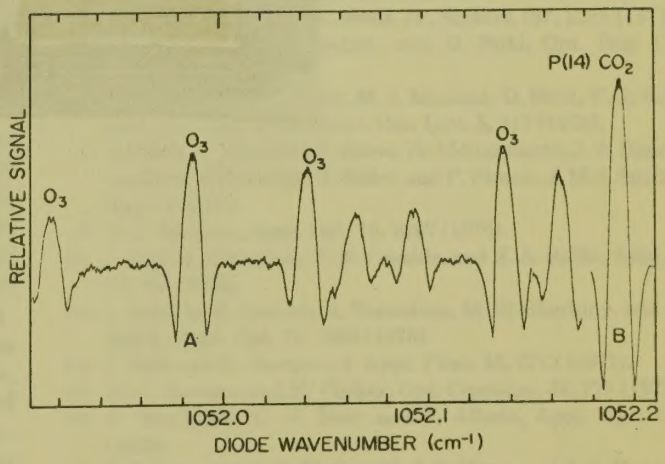


Fig. 5. Detection of atmospheric ozone. This second harmonic scan was taken over the same wavenumber region as in Fig. 4 and employed the same pathlength and total pressure. A 1-sec time constant was used. The additional lines are discussed in the text. The estimated ozone concentration was ~38 ppb. Note that lines A and B are repeated in Fig. 6 but in a different scale. Note also that the instrumental sensitivity was reduced by a factor of 10 for line B.

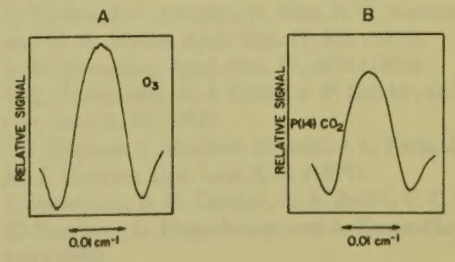
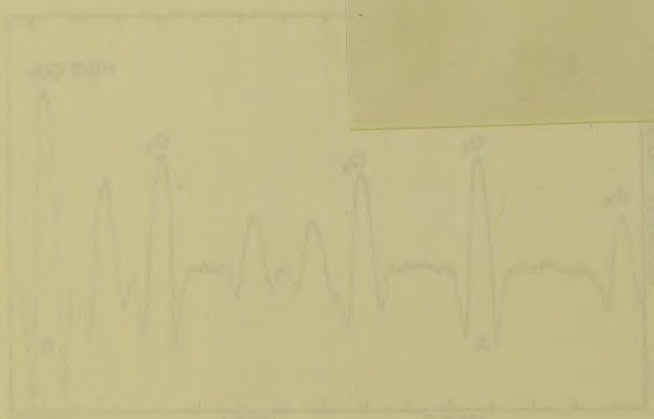


Fig. 6. Selected lines from Fig. 5 taken under the same conditions, except that the instrumental sensitivity for B was a factor of 20 lower than A. The absorption coefficient for the central ozone line is ~5 $\times 10^{-6}$ m^{-1}.



$8_{35} \rightarrow 9_{54}$ line of HDO, listed at 1052.144 cm^{-1} on the AFCRL compilation.[25] The remaining two lines are caused by some unidentified constituent of the air above our laboratory. Figure 5 clearly illustrates one of the problems of using CO_2 lasers for differential absorption measurements to monitor atmospheric ozone. If the $P(14)$ CO_2 line is used as a probe, most of the atmospheric absorption of the laser beam will be caused by the atmospheric CO_2, with only a small contribution from ozone. There can also be additional interferences from HDO, and possibly from the unknown lines observed near 1052.0 cm^{-1}. Clearly, a complicated calibration procedure is required to account for such interferences. Our LAS does not suffer from such complications, as the tuning ability of the diode laser enables us to concentrate on those ozone lines that are free from interferences. For example, to monitor ozone in the air, it is sufficient to scan over one isolated ozone line, as shown in Fig 6(A). This scan is taken with a 3-sec time constant to improve the SNR. For comparison, we show in Fig. 6(B) the $P(14)$ CO_2 line as observed in air. For the latter, the instrumental sensitivity was reduced by a factor of 20. These lines correspond to A and B in Fig. 5.

We made no attempt to calibrate accurately the ozone concentrations observed in Figs. 5 and 6 nor to ensure that our samples of air remained uncontaminated by nearby vehicle and air-conditioner exhausts. These scans were simply taken to illustrate the LAS technique and sensitivity. However, one can obtain a reasonable estimate of the ozone concentration by comparing the measured absorption with the known line strength of the observed ozone lines.[8] This procedure gives an ozone concentration of 35 ppb. As a check on this calculation, we compare the measured ozone absorptions directly with the absorption observed on the $P(14)$ CO_2 line. If the atmospheric CO_2 concentration is assumed to be 330 ppm,[25] we estimate that the ozone concentration is 38 ppb, in good agreement with the previous calculation. It follows that the noise level in Fig. 6 corresponds to ~0.5-ppb ozone in air. These measurements employed a 5-m long White cell. Similar sensitivities have been obtained using a portable commercial 0.75-m long White cell.[11] We are in the process of developing a fully portable instrument that will have a sensitivity of better than 1 ppb of ozone in both ground-based and airborne applications.

V. Conclusions

In this paper we report the accurate frequency measurement of over 100 lines in the ν_3 band of ozone. These measurements were made with a tunable diode laser and a temperature-stabilized germanium etalon. Despite the relative simplicity of the technique, we achieve an accuracy of better than 10 MHz in our measurements. This accuracy compares well with that achieved by the more complicated heterodyne techniques of high-resolution spectroscopy. In our measurements we have primarily concentrated on those ozone lines that are close to CO_2 laser lines. Finally, we have demonstrated the use of a tunable diode laser for point monitoring of ambient concentrations of ozone at a sensitivity of 0.5 ppb.

This work was supported by the Department of National Health & Welfare, Environment Canada, Ontario Ministry of the Environment, and Atmospheric Service, Environment Canada.

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APPENDIX 7-1

October 1979

WORLD HEALTH ORGANIZATION

PILOT PROJECT ON THE ASSESSMENT OF HUMAN EXPOSURE TO AIR POLLUTION

History

The framework of the W.H.O. Environmental Health Monitoring Programme was developed during a meeting of interested scientists in Geneva in July 1974. The objectives of this programme are:

(1) technical cooperation with Member States to establish and develop local or national environmental health monitoring systems;

(2) international exchange and use of information for assessment and improvement of environmental conditions related to public health;

(3) Contribution to the Global Environmental Monitoring System and other Earthwatch components related to human health.

In the context of this framework a UNEP/WHO group of government experts on Health-Related Monitoring met in Geneva in March 1977 to provide UNEP and WHO with advice on how to integrate current monitoring activities, and supplement them with additional ones that would make possible the assessment of human exposure to pollutants. Their recommendations were:

(1) to undertake a pilot project in a selected number of urban areas to estimate human exposure and percent population at risk due to exposure to a number of pollutants;

(2) to include developing countries, since their air pollution problems might be different from industrial countries, requiring a different approach to monitoring;

(3) to pay particular attention to indoor vs outdoor air quality relationships.

Out of these guidelines a project document was drafted, which sketched the outline of the proposed study in some detail, including the financial aspects. This was formally described as "United Nations Environment Programme Pilot Project on the Assessment of Human Exposure to Air Pollutants. Project # FP/1301-78-03. Priority area: GEMS. Scope: Global. Implemented by WHO, supported by WMO". The date of this project document was August, 1978. At the same time, Dr. J. Mahoney of Harvard University was asked to write a draft workplan in the form of a background paper, which was published in February, 1979.

In February, 1979, a meeting was held in London, England, at the MRC Toxicology Unit, Clinical Section, St. Bartholomew's Hospital Medical College. Experts gathered there for the purposes of consultation included B. Ferris, P. Lawther and R. Waller, along with ten other scientists interested in environmental health. The objective of the meeting was to give advice on the approaches to be taken to implement the project. The group used Mahoney's background paper (prepared specifically for this purpose) as guidance, and made statements on ten specific areas of concern:

(i) Preliminary studies
(ii) Monitoring network design
(iii) Indoor/Outdoor studies
(iv) Distribution of I/O and personal samplers
(v) Diary of activities
(vi) Meteorological data
(vii) Measurement principles and techniques
(viii) Interface with biological monitoring
(ix) Demographic and socio-economic data
(x) Quality assurance.

Their statements on these areas agreed in many respects with those of the background paper, although "considering the complexity of the project (they) cautioned to adopt the simplest approach possible". In parallel with these developments, the McMaster school children study was beginning to get under way.

During August 1978, Dr. Pengelly and Dr. F. Silverman (Gage Research Institute, Toronto were invited to a meeting with W.H.O. representatives

in Toronto by Dr. T. Munn, who is a member of the W.M.O. and one of the consultants to the UNEP/WHO study. There, Dr. Pengelly presented the results of the 1977 Pilot Study, and Dr. Silverman outlined her work and a proposed study of asthmatics in both studies,an examination of indoor air quality was vital.

Since that time, Dr. Munn, on behalf of the Toronto-Hamilton group (THG), and Dr. H. de Koning, the Project secretary, have been communicating regularly. As well, a proposal that the THG be designated a participating centre has been submitted to WHO/UNEP. It appears that they view this participation with interest. A special meeting involving WHO/UNEP personnel and representatives of the THG has been called for the second week of November (1979) in Toronto, Canada.

APPENDIX 7-2

WORK PLAN FOR THE TORONTO/HAMILTON COMPONENT OF WHO/UNEP PROJECT NO. FP/1301-78-03

PILOT PROJECT ON THE ASSESSMENT OF HUMAN EXPOSURE TO AIR POLLUTANTS

Preamble: This document describes a Work Plan for the Toronto-Hamilton component of the WHO/UNEP Pilot Project on the Assessment of Human Exposure to Air Pollutants. The Work Plan was developed at a Consultation held November 7-9, 1979 in Toronto, Canada. The Co-Chairmen of the Consultation were Dr. R. E. Munn (IES, Toronto) and Dr. H. de Koning (WHO, Geneva), and the participants are listed in Appendix 1.

1. The WHO/UNEP Project

The Project is described in the following two documents available from WHO, Geneva:

1. Report of a consultation on the pilot project on the assessment of human exposure to air pollutants, EHE/ETS/79.5, March 1979, 7 pp.
2. Background paper for a pilot project on the assessment of human exposure to air pollutants (prepared by Env. Res. and Tech. Inc., Concord, Mass. 01742, U.S.A., Feb. 1979, 49 pp.)

The main objectives of the Project are:

1. to assess the air pollution exposures (both indoor and outdoor) of samples of human populations in 5 or 6 urban areas;
2. to attempt to relate these exposures to air quality as measured at standard outdoor networks of monitoring stations.

To simplify the task, dwellings where there are significant indoor sources of pollution (smokers, gas stoves, etc.) should not be monitored within the Project. In the case of personal monitors, the volunteers should not be subject to unusual occupational exposures.

2. Time Schedule for the Toronto/Hamilton Component

Phase 0 (preprogramming):	Nov. 15/79 - Mar. 31/80
Phase 1 (pilot studies):	Apr. 1/80 - Jul. 31/80
Phase 2 (operational):	Aug. 1/80 - Jul. 31/81
Phase 3 (final analysis and report writing)	Aug. 1/81 - Dec. 31/81

Phases 0 and 1 are already approved by the WHO. Phases 2 and 3 will be considered at a meeting of the International Advisory Committee in July, 1980.

3. Project Control

For convenience, the WHO Contract will be with the University of Toronto, with Dr. R. E. Munn (IES) listed as principal investigator.

A local Steering Committee is established, with representatives from the University of Toronto, McMaster University, the Ontario Ministry of the Environment, Health and Welfare Canada, and the Atmospheric Environment Service. The Committee will meet once every two months.

An international Advisory Committee is established, with representatives selected by WHO. It met in November, 1979 and will meet again in July 1980, and possibly in December, 1981.

4. Programme

4.1. Phase 0

Both the University of Toronto and the WHO will seek to provide suitable indoor/outdoor monitors for the Project. A decision will be made by February 15, 1980 on the monitors to be used.

The pollutants to be monitored will be suspended particulates and SO_2. A 24-hour sampling period for suspended particulates will be used for indoor/outdoor monitors (in order to make comparisons with MOE high volume sampler measurements).

4.2. Phase 1

The indoor/outdoor monitors will be inter-compared indoors with the U. of T. personal monitors and outdoors with standard MOE air quality monitors.

Criteria will be developed for (a), siting of indoor monitors,* and (b), selection of volunteers for personal samplers. Quality control protocols and operational schedules for Phase 2 will also be formulated.

4.3. Phase 2

(A) Equipment

(1) U. of T.

In addition to the two personal monitors to be provided within resources, the U. of T. will require 4 samplers, (2 for indoors, 2 for outdoors) to be supplied through the WHO Contract.

(2) McMaster

McMaster will require 8 samplers (4 for indoors and 4 for outdoors) to be provided through the WHO contract. These samplers should be identical with those used in Toronto.

* In the McMaster case, this will include field studies to determine representative sites in each of the schools chosen.

(3) Spares

The WHO will provide 4 spare indoor/outdoor samplers to be shared by the Toronto and McMaster groups. Depending on costs and ease of calibration, the WHO may also provide one spare personal sampler.

(B) Schedule

(1) U. of T.

From its lists of asthmatics and controls, the Gage Research Institute will seek the cooperation of a few volunteers in houses where there are no smokers or gas stoves. Each volunteer will be recruited for 2 weeks twice a year for a programme of personal, indoor and outdoor daily sampling of suspended particulates and SO_2. Two such persons (from different homes) will be asked to volunteer each week, which means that 2 X 13 homes will be sampled during Phase 2, once in the heating season and once in the non-heating season. This is the maximum programme envisaged, but there is no doubt that the resulting data set will contain gaps.

From the point of view of the WHO, the main interest is in the control group rather than in the asthmatics. At least half of the volunteers should be non-asthmatics, and should be chosen from various types of homes and apartments (with respect to age, location, heating system, air conditioning, etc.).

Where a volunteer spends a significant fraction of the week in an office, store or factory, efforts should be made to sample that environment also (perhaps using one of the spare instruments).

The outdoor sampler will be located at the nearest MOE monitoring station.

(2) McMaster

The Board of Education for the City of Hamilton will be approached for permission to set up indoor and outdoor air quality monitors at four schools, one to be chosen from each of the four sectors into which the city has been divided. These schools will be chosen for their proximity to existing particulate and sulphur dioxide monitoring stations operated by the Ontario Ministry of the Environment (OME) or by McMaster University. Additional information for relating WHO sampler readings to Particle Size Distribution may also be available coincidentally, depending upon the locations of the schools chosen. It should be noted that locations chosen for McMaster's particulate samplers and sulphur dioxide samplers are not the same; siting requirements differ greatly.

The McMaster Group is hoping to run the four pairs of indoor-outdoor monitors every second day during Phase 2. In addition, an increase in the sampling frequency for particulates is planned, from every sixth day to every second day. This will only apply to the McMaster University sites nearest the chosen schools, not to the OME sites, if one of these should be closest.

It will be possible to relate sulphur dioxide levels as measured by the WHO supplied samplers to levels provided by the nearest McMaster samplers for only six weeks out of every 24, for any given school, due to the schedule of rotation of the samplers involved. However, continuous data will be available from these samplers during that six week period.

(3) Ministry of the Environment -

The outdoor samplers will be co-located at MOE monitoring stations. In the case of suspended particulates, MOE will operate daily whenever Project samplers are running.

4.4. Phase 3

A five-month period is planned for final analysis and report writing.

5. Manpower

5.1. U. of T.

One technician and one engineer are required to complete development of the personal monitors, and to adapt, validate and build duplicates for use as indoor/outdoor monitors. These persons are also needed to carry out the field programme, to develop quality control procedures, to analyze data and to assist in the preparation of final reports (funding to be requested from MOE and/or Health and Welfare).

One additional technician is required to assist in the health-related part of the field programme, including personal monitoring (required by WHO) and health effects, diary of activities, etc.(not part of the WHO Project) (funding to be requested from MOE and/or Health and Welfare).

One half-time project coordinator is required to coordinate the programme, including management of the field work, data collection and management, data analysis and preparation of final reports (funding to be requested from MOE and/or Health and Welfare).

5.2. McMaster

One technician, in addition to the present staff, will be required to carry out this project. (funding to be requested from OMOE and/or Health and Welfare Canada). This person's duties would include assisting in calibration and procedural development in co-operation with the Gage Research Institute, carrying out the field programme (including quality control), analyzing data, and assisting in th preparation of final reports.

6. Preparation of a Monograph on the Design of Health-Related Monitoring Systems

The designated Principal Investigator, R. E. Munn, has agreed to submit to the WHO by April 1, 1980, the first draft of a monograph on the design of health-related monitoring systems. The manuscript will be reviewed by a small group of experts to be arranged by the WHO in the summer of 1980. (This monograph is already in preparation for the Rockefeller Foundation, and will not involve additional commitments of time or money except for the cost of the Review Committee meeting, plus photocopying and other incidental expenses.)

7. Additional Related Studies

Although not part of the WHO Project, the following activities would be valuable adjuncts.

(a) The health effects studies at Toronto and McMaster are closely linked to the WHO/UNEP Project although not components. There is no doubt that the scientific content and relevance of both the Project and the health studies will be enriched by their concurrency.

(b) The AES will seek approval to undertake or support a concurrent indoor/outdoor sulphate monitoring study, capitalizing on existing programmes within the WHO/UNEP Project. Health and Welfare Canada will seek approval to undertake or support a concurrent sulphate health-effects study.

(c) IES will seek support for a graduate student in the social sciences to undertake a life style analysis (with respect to air pollution exposures), seeking to improve existing criteria for selecting volunteers and dwellings, and seeking to generalize the data collected within the Project.

(d) NO_x and/or oxidant measurements could usefully be made concurrently during Phase 2 of the Project.

(e) A review of the engineering literature on building ventilation rates would be valuable.

(f) A field study should be undertaken of the variability in air quality in various rooms of the dwellings being monitored.

(g) Using an instrumented trailer, it would be desirable to undertake indoor/outdoor air quality relations, where the outdoor monitor is immediately adjacent to the structure.

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